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Remnants of Early Archean Impact Deposits on Earth: Search for a Meteoritic Component in the BARB5 and CT3 Drill Cores (Barberton Greenstone Belt, South Africa)

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Abstract

The first 2.5 billion years of the terrestrial impact history are not documented by any impact structures. Only a few spherule layers of impact origin are known, most of them of late Archean to early Proterozoic age. In the Barberton Greenstone Belt (South Africa), several spherule horizons (layers S1 to S4, possibly up to S8, with ages between ~3.5 and ~3.2 Ga) are amongst the oldest deposits from large bolide impacts onto Earth. Impact evidence is limited to (highly) elevated siderophile element contents and Cr isotopic compositions. Other isotope tools, such as the ^{187}Re-^{188}Os radionuclide system in combination with high-precision concentration data for siderophile elements, might be useful to confirm the propositions regarding the presence of meteoritic components made so far. Two recently recovered drill cores from the central and northern Barberton area (CT3 and BARB5) with as many as 18 spherule layer intersections of Paleoarchean age (some of which may be due to tectonic duplication, some might correlate with the S2 to S4 layers) provide an outstanding opportunity to gain new insight into the early impact bombardment of Earth.

We present new mineralogical, chemical, and ^{187}Re-^{188}Os isotope data on CT3 and BARB5 drill core samples. Spherules in most layers exhibit undeformed shapes and include vesicles. Sulfides frequently are present in both matrix and spherules. Osmium data reveal a trend between the spherule-free horizons (intercalating the spherule layers) and spherule-matrix aggregates. Whereas the former typically exhibit elevated $^{187}Os/^{188}Os$ ratios of up to ~1.2 and low Os and Ir concentrations below several hundred ppt, spherule-matrix aggregates tend to be less radiogenic (down to subchondritic present day $^{187}Os/^{188}Os$ ratios) with Os and Ir concentrations as high as in chondrites. Chromium-Ir correlations for CT3 and BARB5 samples mirror earlier results on S1 to S4 layers and can be interpreted in favor of an impact origin of the here investigated spherule horizons.

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Keywords: Barberton Greenstone Belt; Archean impacts; spherules; highly siderophile elements; Osmium isotopes

1. Introduction

The oldest known impact structure on Earth is about 2 billion years old. Considering an age of the Earth of just over 4.5 billion years, this means that the impact record for more than half of the Earth's history is missing. In the absence of any conclusive impact record in the oldest rocks on Earth, we need to look at "younger" rocks. The first "real" rock record of impact events dates to about 400 to 500 million years after the end of the Late Heavy Bombardment (LHB; see next section), in the form of several Neo-Archean (distal?) ejecta layers (see review by, e.g., Glass and Simonson [1]). Four distinct spherule horizons in the Barberton Greenstone belt, South Africa (designated S1 to S4), with ages between about

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3.5 and 3.2 Ga, have been proposed as being of impact origin (e.g., Lowe et al. [2]). The spherules are mostly spherical particles, up to a few mm across, of quenched melt droplets that supposedly formed by condensation from impact vapor clouds. The spherule layers are coarse-grained and have been interpreted to reflect high-energy depositional events in otherwise low-energy, quiet water environments. The original mineralogical and chemical composition of the spherules has been almost completely changed by alteration. The stratigraphic positions of these layers at various geographic locations are difficult to correlate and the possibility exists that some of the layers represent duplication. These spherule layers show extreme enrichments in the PGEs (in some samples exceeding the PGE abundances found in chondritic meteorites up to four times), unlike modern impact ejecta deposits (for example, those at the Cretaceous-Tertiary boundary, or in the late Eocene), which has caused some questions regarding the initial impact interpretation.

Spherule layers from the Barberton Greenstone Belt (see Table 1a for a compilation) have been interpreted as the result of large asteroid or comet impacts onto the early Earth (e.g., Lowe et al. [2]). The extreme enrichments in the PGEs and other inconsistencies caused Koeberl and Reimold [3] to question the impact interpretation. The correlation between the abundances of iridium and arsenic, a very mobile element, in samples from the Barberton spherule layers, all of which have been subject to pervasive transformation into secondary mineral assemblages, indicates remobilization of both elements; this infers that the PGE signature in these samples might not be primary (e.g., Koeberl and Reimold [3]; Reimold et al. [4]). Kyte et al. [5] reported chromium isotopic anomalies in samples from several of these layers that support the presence of an extraterrestrial component. A comprehensive study of sedimentary, petrographic, mineralogical, and geochemical characteristics from a set of new samples of spherule layers discovered between 510 and 512 m depth in the 760-m-long ICDP drill core BARB 5 from the Barite Valley Syncline, as well as samples from the CT3 location, of the Barberton Greenstone Belt (see Table 1b) has been carried out.

Table 1a: Comparative data on Archean impact spherule layers (after compilation in Reimold and Koeberl [6])

Name	Geographic Location [A]	Host formation	Group	Age [B]	Evidence for an impact origin [C]
n.a.	HB, Western Australia	Wittenoom	Hamersley	2.54	SM, Sph(qt,rv), Ir, PGE
n.a.	GWB, South Africa	Reivilo	Ghaap	2.56	Sph(qt,rv), NiSp, Ir, PGE
n.a.	HB, Western Australia	Carawine	Hamersley	2.63	SM, Sph(qt,rv), Cr, Ir,
n.a.	HB, Western Australia	Jeerinah	Fortescue	2.63	SM, Sph(qt,rv), Cr, Ir, PGE
n.a.	GWB, South Africa	Monteville	Ghaap	2.60 - 2.65	SM, Sph(qt,rv), Ir, PGE
S1	PC, Western Australia	Apex Basalt	Warrawoona	3.472	Sph(qt)
S1	BGB, South Africa	Hoggenoeg	Onverwacht	3.472	Sph(qt,rv), Ir(?)
S2	BGB, South Africa	Mendon	OW/FT	3.256	Sph(qt), Ir(?), Cr
S3	BGB, South Africa	Mapepe / Ulundi	Fig Tree	3.243	Sph(qt,rv), NiSp, Ir, Cr
S4	BGB, South Africa	Mapepe	Fig Tree	3.243	Sph(qt?,rv), Ir, Cr
S5	BGB, South Africa	Belvue Road	Fig Tree	3.225	Sph
S6	BGB, South Africa	Mendon	Onverwacht	3.308	Sph
S7	BGB, South Africa	HG/KR	Onverwacht	3.416	Sph
S8	BGB, South Africa	Mendon	Warrawoona	3.298	Sph

[A] For detailed type localities see references in Reimold and Koeberl [6]. [B] Ages in Ga; [C] Cr = Cr isotope data, Sph = altered spherules, Sph(qt) = altered spherules with quench tecxtures, Sph(rv) = altered spherules with relict vesicles, SM = shock metamorphosed rock, PGE = platinum group elements, Ir = Ir anomaly, Cr = Cr isotope data, NiSp = Ni-rich spinels; n.a. = not applicable; n.d. = not determined; HB = Hamersley Basin; GWB = Griqualand West Basin; BGB = Barberton Greenstone Belt; PC = Pilbara Craton; OW/FT = Transition zone from Onverwacht to Fig Tree; HG/KR = Transition zone from Hoggenoeg to Kromberg; For references, see Koeberl and Reimold [6].

2. Methods

Samples (see details in next section) were powdered for analysis. About 50 to 300 mg sample were spiked with a mixed tracer composed of ^{185}Re and ^{190}Os isotopes and digested in 7 ml inverse aqua regia (HNO_3-HCl: 5+2 ml) at 250°C and 125 bar pressure in an Anton-Paar high pressure asher, followed by solvent extraction (Cohen and Waters [7]) and microdistillation purification (Birck et al. [8]) for Os and anion exchange chemistry for Re. Osmium isotope ratio measurements were carried out using a Triton Thermal Ionization Mass Spectrometer at the Department of Lithospheric Research at the University of Vienna, Austria (via a peak hopping measurement sequence using the Triton SEM detector or using Faraday Cups). Corrections for oxide interferences, ^{187}Re isobaric interferences on ^{187}Os, and mass fractionation were conducted offline. The Os total processing blank was 0.5 ± 2 pg (n = 5) and, therefore, insignificant for most samples. Repeated N-TIMS measurements of 0.1 to 1 ng loads of the *D*urham *R*omil *Os*mium *S*tandard (DROsS) standard solution were performed and replicate analyses made of international reference materials and rock powders of known composition (e.g., TDB-1, OKUM). All values are within the 2 sigma uncertainty of average values as reported in the literature. Rhenium was measured using a Thermo Element ICP-MS in single collector mode at the Steinmann Institute at University Bonn. Total blanks for this study (n = 5) were 5-15 pg for Re. Chromium and Ir abundances (as well as those of about 30 other minor and trace elements; not discussed here) were determined by Instrumental Neutron Activation Analysis (INAA) after irradiation at the Triga Mark-II reactor at the Atomic Institute of the Austrian Universities.

Table 1b: Comparative data on recently recovered BGB drill-core sections, containing spherule layers of unknown relation to the S1 to S4 layers.

Name	Geographic Location [A]	Host formation	Group	Age [B]	Evidence for an impact origin [C]	References
CT3	BGB, South Africa	Bien Venue	Fig Tree	3.26-3.23	PGE(?)	[9, 10]
BARB5	BGB, South Africa	Mapepe	Fig Tree	3.26-3.23	NiSp, PGE(?)	[11-15]

The CT3 core section includes 17 spherule horizons over an interval of 150 m, whereas BARB5 represents a 22 cm thick core interval containing 4 spherule horizons. It is not known, whether these spherule horizons can be related to previously described spherule beds from the BGB (see Table 1a) or if they represent different units (and if so, how many horizons may result from tectonic duplications).

3. Samples and Results

In the cause of two recent drilling projects, the number of known spherule layers from the Barberton area was significantly increased. An International Scientific Drilling Program (ICDP) project in the Barite Valley Syncline in the north-central BGB recovered the BARB5 drill core with a length of 760 m. Four new spherule layers, each about 4 cm thick, were identified in the core interval between 511.29 and 511.51 m depth, all separated by shale and chert within the 3.26 to 3.23 Ga old middle Mapepe Formation of the Fig Tree Group (e.g., Hoehnel et al. [10]). All spherules exhibit sizes between 0.3 and 2 mm; size gradation is not obvious. Stratigraphically, these spherule layers may belong to the same interval as the previously studied S3/ S4 layers (ibid). The numbers utilized for the BARB5 drill core samples analyzed in this study roughly correlate with an increasing spherule to matrix ratio. While sample B2 is devoid of any spherules, matrix components tend to decrease from B-5 to B-15. This correlates with the Cr, Ir, Os, and Re contents, which are lowest in the B2 sample that also exhibits the most radiogenic $^{187}Os/^{188}Os$ composition (~0.3; see Table 2a).

However, due to possible Os and Ir (or generally speaking: compositional) heterogeneities within the sampled spherule horizons from the BARB5 core section, the (only rough) increase in spherule-matrix ratios observed in the hand specimen is not strictly mirrored in the Os and Ir concentrations. Present day $^{187}Os/^{188}Os$ ratios are in part subchondritic for the spherule horizons (~0.106 to ~0.116 for samples B-8 and B-15) but values back-calculated to ~3.4 Ga are indistinguishable from the chondritic $^{187}Os/^{188}Os$ evolution line (~0.105 to ~0.112 for samples B-8 and B-15 compared to ~0.105 for chondrites at ~3.4 Ga), giving so far no support for subchondritic initial values as reported by Morel et al. [16] for samples from the S3 and S4 layers. However, possible Re loss during hydrothermal alteration or metamorphic overprint may obscure the real initial values in these samples, a fact that might also explain the subchondritic $^{187}Re/^{188}Os$ ratios in some of the analyzed samples.

The CT3 drill core is from the northern BGB and contains not less than 17 spherule layers over a stratigraphic interval of 150 m, occurring along the transition zone between the Onverwacht and Fig Tree groups (Hoehnel et al. [10]). It is far from clear whether some of these spherule layers might actually represent tectonic duplication. Tectonic duplication of spherule layers even on a centimeter to decimeter scale has been reported for other drill core sections from the Barberton area (e.g., Reimold et al. [4]). It also has to be evaluated whether the CT3 layer(s) is/are correlated to the S2 layer, which occurs in the same stratigraphic unit.

Samples 523i and 522f represent spherule-fee intercalations of country rock, whereas all other samples from the CT3 core sample spherule horizons from varying depths (see Table 2b for details). This is mirrored in the Ir and Os contents, which are comparably low in the country rocks, ranging from ~0.12 to 0.97 ppb for Ir and ~0.25 to 1.3 ppb for Os (which is still higher than average modern continental crust), and are elevated in the spherule horizons (between ~6 and 2068 ppb in Ir and ~3 ppb and ~4310 ppb for Os, respectively). Chromium concentrations also exhibit the lowest values in the spherule-free shale and chert intercalations (see Table 2b). $^{187}Os/^{188}Os$ and $^{187}Re/^{188}Os$ ratios also significantly vary between the country rocks and spherule horizons (from 0.21 to 1.13 for $^{187}Os/^{188}Os$ and ~4.5 to 99.6 for $^{187}Re/^{188}Os$ ratios in country rocks compared to 0.11 to 0.17 for $^{187}Os/^{188}Os$ and ~0.06 to ~0.33 for $^{187}Re/^{188}Os$ ratios in the spherule horizons). Back-calculated $^{187}Os/^{188}Os$ ratios for all samples are superchondritic.

Table 2a: Selected element abundances and Re-Os isotope data for samples from the BARB-5 drill core

sample	Cr	Ir	Os	Re	$^{187}Os/^{188}Os$	$^{187}Re/^{188}Os$
B-2	82.2	0.91	8.6	0.337	0.3041 (32)	0.191 (6)
B-5	736	8.4	10.8	0.234	0.1344 (3)	0.103 (3)
B-7	3561	47.5	57.1	3.192	0.1245 (15)	0.266 (8)
B-8	1596	321	944	3.411	0.10644 (1)	0.017 (1)
B-12	2196	386	396.1	26.14	0.12130 (3)	0.314 (9)
B-13	434	35.8	42.57	2.529	0.1691 (40)	0.300 (9)
B-14	1772	166	150.5	4.629	0.1256 (6)	0.144 (4)
B-15	3211	671	503.2	5.877	0.11583 (9)	0.056 (2)

Concentrations for Ir, Os, and Re in ppb, for Cr in ppm. Chromium and Ir data are by INAA; Os and Re by isotope dilution.

Table 2b: Selected elemental abundances and Re-Os isotope data for samples from the CT3 drill core

Sample	Depth	Description	Cr	Ir	Os	Re	$^{187}Os/^{188}Os$	$^{187}Re/^{188}Os$
432o	65.45 m	Shale + Sph + sulfides	6862	2068	4312	57.39	0.11374 (30)	0.063 (2)
511c	66.37 m	Sph + sulfides	753	8.68	2.92	0.189	0.1668 (12)	0.310 (9)
522f	66.61 m	Shale	90.4	0.97	0.25	4.666	1.1265 (90)	99.60 (3)
522g	66.64 m	Sph	449	6.12	2.94	0.206	0.13380 (42)	0.334 (9)
523i	66.68 m	Chert	20.7	0.12	1.30	0.109	0.21386 (22)	4.544 (8)

All concentrations in ppb, except Cr (ppm). Chromium and Ir data by INAA; Os and Re by isotope dilution.
Sph = altered spherules

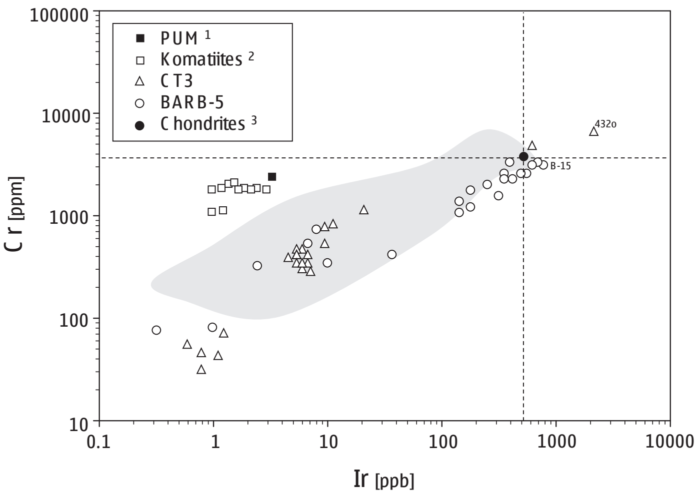

Fig. 1. Plot of Ir versus Cr abundances in BGB spherule beds, BGB komatiites, primitive upper mantle (PUM), and CI chondrites. The shaded area represents literature data for the S1-S4 layers (Lowe et al. [2] and references therein). The BGB country rocks and spherule beds from recently recovered drill cores, including those of the here investigated CT3 and BARB5 samples, lie on a trend intersecting CI chondrites. Samples from the BGB spherule layers are distinct from the primitive mantle and komatiites. 1 = Palme and O'Neill [17], 2 = Puchtel et al. [18], 3 = Palme and Jones [19].

4. Discussion

Figure 1 displays a correlation between the abundances of Cr and Ir in spherule layers S1 to S4 from the BGB (shaded area). This correlation allows a clear discrimination between komatiites and spherule layer samples, the latter defining a trend intersecting chondritic meteorites. This trend, therefore, represents one of the key arguments for a possible impact origin of BGB spherule beds. Lowe et al. [2] concluded that the highest Cr and Ir enrichments, measured in the S1 and S2 spherule beds, clearly point toward a chondritic contamination, whereas the S3 and S4 beds are dispersed at lower Cr and Ir values. Notably, significant meteoritic components were reported for S2 to S4 layers based on Cr isotope investigations (e.g., Kyte et al. [5]), exemplifying that highly subchondritic Cr and Ir abundances do not preclude an extraterrestrial origin. Our present data from BARB5 and CT3 exactly match the trend defined by layers S1 to S4 in the Ir vs. Cr space. Few samples from both drill cores exhibit superchondritic values, exceeding the highest concentrations measured for the S1 to S4 layers (e.g., Reimold et al. [4] and Lowe et al. [2], and references therein). However, chondrites exhibit a wide range of Ir values from ~470 ppb in CI type chondrites to ~740 ppb in CO and CV type chondrites. These values cover all measurements from this study, except sample 432o from the CT3 layer, exhibiting an Ir concentration of ~2.1 ppm. The common trends of all BGB spherule layer samples (including spherule-free chert and shale intercalations), therefore, provide strong support for a meteoritic contamination within both layers, especially in the spherule-containing samples 432o (CT3) and B-15 (BARB5). However, it is currently not clear whether the here investigated BARB5 and CT3 spherule beds are correlated to the S3 or S4 layers and/or the S2 layer, or if they represent one or more different units.

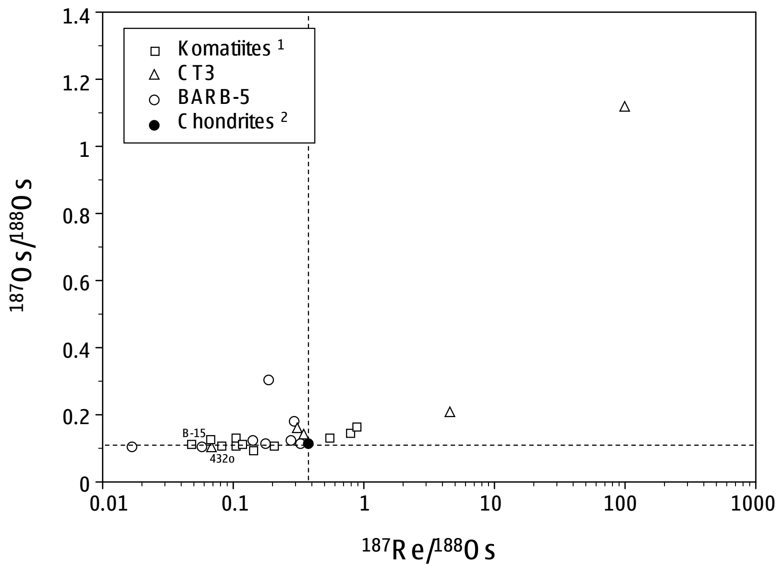

Fig. 2. Plot of $^{187}Re/^{188}Os$ versus $^{187}Os/^{188}Os$ isotopic ratios in BARB5 and CT3 samples. Also shown are data for komatiites and CI chondrites. No data are available for layers S1 to S4. The BARB5 and CT3 samples lie on a trend typical for komatiites. Drill core samples suspected to contain a meteoritic component (432o from CT3 and B-15 from BARB5; see Fig. 1) exhibit subchondritic $^{187}Re/^{188}Os$ ratios. 1 = Puchtel et al. [18], 2 = Shirey and Walker [22].

The ^{187}Re-^{187}Os isotope system is based on the formation of ^{187}Os by β^- decay of ^{187}Re (half-live = 42.3 ± 1.3 Ga). As a result of the different geochemical behavior of Re and Os (Re behaves more incompatible compared to Os during melting events), $^{187}Os/^{188}Os$ isotope and Re/Os elemental ratios of crustal rocks are typically higher compared to the possible impactor material, an effect that is all the more pronounced the older and the more felsic the target material is. This was extensively used in the past to decipher the presence of even very small amounts of meteoritic contaminations in impact melt rocks from a variety of impact locations (down to the sub-percent level; see Koeberl [20] and references therein). However, komatiitic volcanics are present throughout the BGB and, therefore, provide a mafic to ultramafic background, limiting the significance of the ^{187}Re-^{187}Os vs. $^{187}Os/^{188}Os$ space used to visualize mixing trends between non-spherule containing country rocks (with elevated Re/Os ratios and radiogenic $^{187}Os/^{188}Os$ ratios, as represented by samples 522f and 523i for the CT3 core as well as sample B-2 for the BARB5 core) and unradiogenic meteorites with lower Re/Os ratios (i.e., chondrites and iron meteorites). Consequently, Fig. 2 does not allow to draw a clear distinction between typical values from komatiites and samples from both drill cores. Furthermore, some samples from both drill cores exhibit slightly subchondritic $^{187}Re/^{188}Os$ ratios, within the range for komatiites (e.g., 432o and B-15, suspected to contain a meteoritic component based on Fig. 1). Komatiites resulted from high degrees of partial melting and therefore extract large proportions of moderately compatible elements from the mantle (e.g. Os), but these values, although high compared to typical crustal rocks, seldom exceed ~2 ppb. A more robust means of identifying meteoritic components and constraining the mass input from impactors based on Os isotopes may thus be provided in the Os vs. $^{187}Os/^{188}Os$ space (Fig. 3). Samples from both drill cores define a trend of significantly less radiogenic values towards higher Os contents. Komatiites are clearly detached from this trend as they all exhibit comparably low $^{187}Os/^{188}Os$ ratios and Os contents. Again samples 432o and B-15 are close to the suggested chondritic end-member.

A special focus has to be given to the back-calculated $^{187}Os/^{188}Os$ ratios for samples from BARB5 (as discussed in the Results section). Samples B-8 and B-15, both elevated in Ir and Os, plot on or slightly above the chondritic evolution line at ~3.4 Ga. This so far did not confirm the findings of Morel et al. [16] on samples from the S3 and S4 layers, who argued for an iron meteoritic impactor in the wake of the subchondritic initial $^{187}Os/^{188}Os$ ratios they calculated for samples from the S3 and S4 layers (which is difficult to be explained by any known chondritic impactor). However, hydrothermal alteration may have caused Re loss in at least some of the samples, possibly explaining the an order of magnitude lower $^{187}Re/^{188}Os$ ratios in samples B-8 and B-15, compared to all other analyzed samples from this drill core (see Table 2a). If so, this could easily lead to subchondritic initial $^{187}Os/^{188}Os$ ratios, which nowadays are obscured because of the possible Re loss. This calls for more detailed investigations on $^{187}Os/^{188}Os$ and $^{187}Re/^{188}Os$ ratios in spherule horizons from the BARB5 drill core and detailed investigations of the extent of hydrothermal activity that has affected these samples.

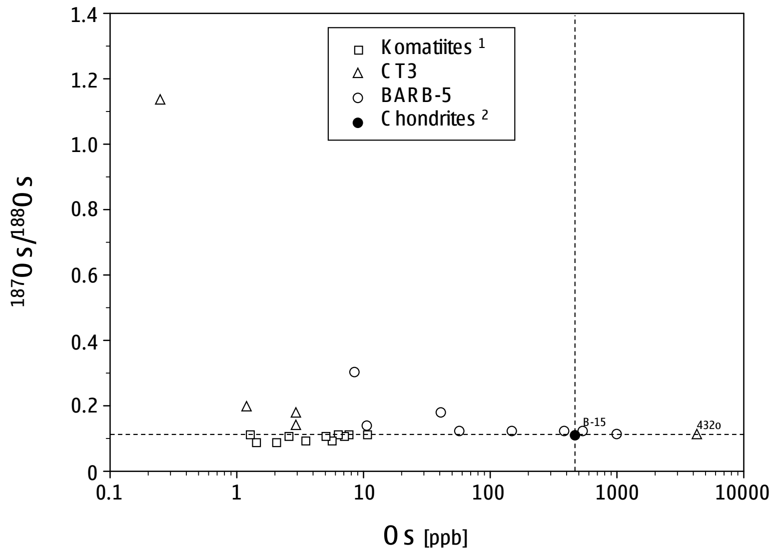

Figure 3: Figure 3: Plot of Os concentrations versus $^{187}Os/^{188}Os$ ratios in BARB5 and CT3 drill core samples. Also shown are data for komatiites (Puchtel et al., 2014) chondrites. BARB5 and CT3 samples lie on a mixing trend between spherule-free country rocks and CI chondrites. Samples B-15 (BARB5) and 432o (CT3) exhibit slightly superchondritic Os concentrations. 1 = Puchtel et al. [18], 2 = Shirey and Walker [22] as well as Palme and Jones [19].

Additional support for the impact hypothesis might come from Os and Ir ratios: chondrite normalized Ir/Os ratios decrease for the CT3 layer from ~2 to ~4 in samples 511c, 522f and 522g towards a value of ~0.5 to ~1 in sample 432o (using CI, CV and CO chondrites as normalizing values). The spherule-free chert sample 523i defines a $(Ir/Os)_n$ value of ~0.09. BARB5 samples vary in $(Ir/Os)_n$ ratios from ~0.1 for the spherule-free end-member towards values ranging from ~0.8 to ~1.4 for the spherule containing samples (B5 to B15, exhibiting varying spherule-matrix ratios). This, however, not necessarily favors the impact hypothesis, as komatiites typically exhibit $(Ir/Os)_n$ ratios around 1 (e.g., Puchtel et al. [18]). Notably, sample 522g from the CT3 core, consisting almost exclusively of spherules, exhibits Cr, Os, and Ir concentrations as well as Os isotope values completely unrelated to any meteoritic contamination. This may point toward heterogeneous distribution of extraterrestrial contaminants within spherules of CT3 layers in close proximity of each other.

5. Summary and Conclusions

Much of the Archean impact record on Earth remains shrouded in secrecy. Large asteroid and cometary bolides have undoubtedly hit the Earth during and in the wake of the late heavy bombardment, but the scars of these impact events have long since been obliterated. However, impact spherules (possibly formed as molten impact ejecta and condensation products from impact plumes or ejecta that were re-melted during atmospheric re-entry) provide information about an impact event even when the source crater cannot be found. Based on their Cr isotope signatures three spherule layers of Paleoarchean age (exclusively preserved in the Barberton Greenstone Belt, South Africa) unambiguously represent the oldest terrestrial impact remnants identified so far. Two recently recovered drill cores from the Barberton area (BARB5 and CT3) with possibly up to 18 new spherule layers of Paleoarchean age provide an outstanding opportunity to gain new insights into the meteorite bombardment of the Early Earth (although some of these spherule layers may be due to tectonic duplication and some might correlate to known impact layers from the Barberton area). We, therefore, are working on a detailed mineralogical and Re-Os isotope investigation on CT3 and BARB5 drill core samples, including concentration data for minor and trace elements. Our preliminary data reveal a strong correlation between the contents of Cr and Ir in all investigated samples from both drill cores, intersecting the chondritic composition and perfectly mirroring trends defined by confirmed impact spherule layers from the Barberton area. Moreover, $^{187}Os/^{188}Os$ ratios vary significantly between samples from the same drill core. Even though spherule-free horizons (intercalating the spherule layers) exhibit radiogenic $^{187}Os/^{188}Os$ ratios (up to ~1.2) and low Os and Ir concentrations (below several hundred ppt), spherule horizons tend to be less radiogenic (down to ~0.106) with Os and Ir concentrations as high as in chondrites. Notably, a significant variation in $^{187}Os/^{188}Os$ ratios and Os as well as Ir concentrations can also be observed within and between the spherule horizons, which, in part, correlate with the spherule content of the analyzed samples (the higher the spherule content, the less radiogenic the Os isotope composition and the higher the Os and Ir concentration). These results can best be explained by contamination of the spherules with meteoritic material derived from an impactor.

Our results for the CT3 and BARB5 cores can, therefore, be interpreted in favor of an impact origin of the investigated spherule horizons. Based on our preliminary data, no definite conclusion can be drawn regarding the nature of the impactor(s), although chondrites might be preferable in the light of the element and isotope trends presented here. A clear assessment of possible tectonic duplications and relations to known impact layers has to be undertaken before a meaningful reappraisal of the Paleoarchean meteorite flux and its possible relation to an extended late heavy bombardment (LHB) [23] can be drawn.

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